

# Memory Effects in Granular Material

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We present a combined experimental and theoretical study of memory effects in vibration-induced compaction of granular materials. In particular, the response of the system to an abrupt change in shaking intensity is measured. At short times after the perturbation a granular analog of aging in glasses is observed. Using a simple two-state model, we are able to explain this short-time response. We also discuss the possibility for the system to obey an approximate pseudo-fluctuation-dissipation theorem relationship and relate our work to earlier experimental and theoretical studies of the problem.

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Granular materials comprise an important class of complex systems whose simple fundamental mechanics gives rise to rich macroscopic phenomenology [1]. Recent experiments on granular compaction [2,3] suggest they are an ideal system for studying jamming, a phenomenon lying outside the domain of conventional statistical physics, yet highly reminiscent of glassiness. These studies showed that a loose packing of glass beads subjected to vertical “tapping” slowly compacts, asymptotically to a higher steady state packing fraction. This “equilibrium” packing fraction is somewhat lower than the random close packing limit,  $\rho_{rcp} \approx 0.64$ , and is a decreasing function of the vibration intensity, typically parameterized by  $\Gamma$ , the peak applied acceleration normalized by gravity,  $g$ . The relaxation dynamics are extremely slow, taking many thousands of taps for the packing fraction,  $\rho$ , to approach its steady state value. During this evolution,  $\rho$  increases logarithmically with the number of taps,  $t$ , which is typical for self-inhibiting processes [4]. The average time scale  $\tau$  of the relaxation decreases with  $\Gamma$ , and in this sense the shaking intensity plays, at least qualitatively, the role of temperature. For small  $\Gamma$ , the relaxation rate becomes so slow that the system cannot reach the steady state density within the experimental time scale. It was also found that compaction can be maximized through an annealing procedure. This process involves a slow “cooling” of the system starting from a high shaking intensity  $\Gamma$ . These slow relaxation and annealing properties of this system are reminiscent of conventional glasses. Another qualitative similarity to glasses is observable in the density fluctuation spectrum of the granular system near equilibrium. The spectrum was found to be strongly non-Lorentzian [3], revealing the existence of multiple time scales in the system. The shortest and the longest relaxation timescales differ by as much as three order of magnitude, and the behavior of the spectrum for the intermediate frequencies is highly non-trivial; in certain regimes it can be fitted with a power law.

These previous experimental observations are suggestive of glassy behavior and this connection has been explored in recent models of compaction using ideas from magnetic systems [5]. However, a more direct test of the

glassy nature of granular compaction comes from measurements of the response of the system to sudden perturbations in the effective temperature, given by  $\Gamma$ . This idea originates from classical experiments for the study of aging in glasses [6] and has recently been explored using computer simulations [7]. In this letter we present direct experimental observations of memory effects in a vibrated granular system obtained by measuring the short-time response to an instantaneous change in tapping acceleration  $\Gamma$  and propose a simple theoretical framework.

We used the experimental set-up described in refs. [2,3]: 1 mm-diameter glass beads were vertically shaken in a tall, evacuated, 19 mm-diameter glass tube, and the packing density of the beads was measured using capacitors mounted at four heights along the column.

The simplest form of this experiment consists of a single instantaneous change of vibration intensity from  $\Gamma_1$  to  $\Gamma_2$  after  $t_0$  taps. For  $\Gamma_2 < \Gamma_1$  (Fig. 1a) we found that on short time scales the compaction rate increases. This is in sharp contrast to what one may expect from the long-time behavior found in previous experiments where the relaxation is slower for smaller vibration accelerations. For  $\Gamma_2 > \Gamma_1$  (Fig. 1b) we found that the system dilates immediately following  $t_0$ .

These results too, are opposite from the long-time behavior seen in previous experiments where the compaction rate increased: Not only does the compaction rate decrease, it becomes negative (i.e. the system dilates). Note that after several taps the “anomalous” dilation ceases and there is a crossover to the “normal” behavior, with the relaxation rate becoming the same as in constant- $\Gamma$  mode. Thus, most of the shaking history is forgotten after a short time.

These data constitute a short-term memory effect: the future evolution of  $\rho$  after time  $t_0$  depends not only on  $\rho(t_0)$ , but also information about the previous tapping history, contained in other “hidden” variables. In order to demonstrate this in a more explicit manner, we modified the above experiment. In this second set of three experiments the system was driven to *the same density*  $\rho_0$  with three different accelerations  $\Gamma_0$ ,  $\Gamma_1$ , and  $\Gamma_2$ . After  $\rho_0$  was achieved at time  $t_0$ , the system was tapped with

the same intensity  $\Gamma_0$  for all three experiments. As seen in Figure 2, the evolution for  $t > t_0$  strongly depends on the pre-history. The need for extra state variables in the problem is consistent with strongly non-Lorentzian behavior of the fluctuation spectrum, observed in earlier experiments [3]. Indeed, if the evolution could be prescribed by a single master equation for local density, it would result in a single-relaxation-time exponential decay of the density fluctuations near equilibrium. Instead, a wide distribution of characteristic times is suggested by the spectrum.

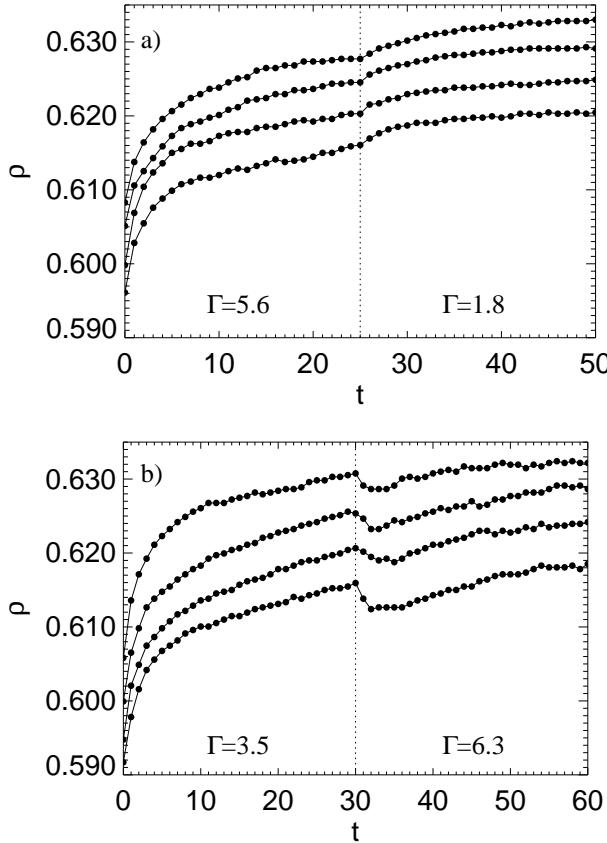


FIG. 1. Evolution of the packing fraction,  $\rho$ , at four heights in the column, as a function of tap number,  $t$ . Two different single-switch experiments: (a)  $\Gamma$  was lowered from 5.6 to 1.8 at  $t_0 = 25$ ; and (b)  $\Gamma$  was increased from 3.5 to 6.3 at  $t_0 = 30$ . Curves are shifted vertically for clarity. Each curve is an average over 4 runs, and the measurement uncertainty in  $\rho$  is  $4 \times 10^{-4}$ .

To give a theoretical interpretation of the above results, we view the problem as an evolution in the space of discrete ‘‘microscopic’’ states corresponding to different realizations of the packing topology (i.e. of the contact network). For each tap there is a possibility for a transition from one microscopic state to another. Since the dynamics is dissipative and the system is under external gravity, a transition to a denser configuration is typically more probable than the reverse one. We now assume that the short-term dynamics of the system are

dominated by a number of local flip-flop modes with relatively high transition rates in both directions.

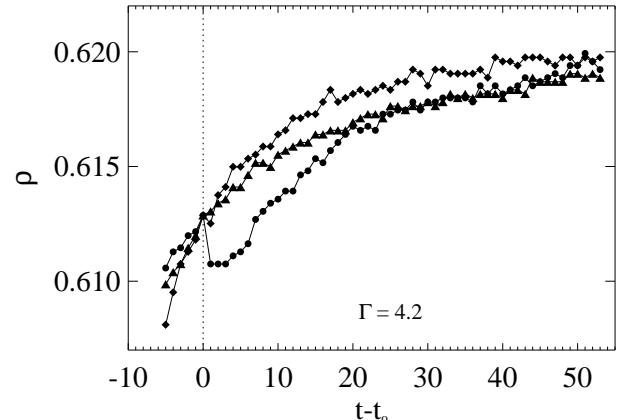


FIG. 2. The time evolution of packing fraction  $\rho$  for a system which was compacted to  $\rho_0 = 0.613$  at time  $t_0$  using three different accelerations:  $\Gamma_1 = 1.8$  (circles),  $\Gamma_0 = 4.2$  (triangles), and  $\Gamma_2 = 6.3$  (diamonds). After the density  $\rho_0$  was achieved, the system was vibrated at acceleration  $\Gamma_0$ . The evolution for  $t > t_0$  depended strongly on the pre-history. Each curve is an average over four experimental runs.

This model replaces the complicated configuration space with a set of independent two-state systems, each of which is characterized by two transition rates,  $\kappa_{e \rightarrow g} > \kappa_{g \rightarrow e}$ .  $\kappa_{e \rightarrow g}/\kappa_{g \rightarrow e}$  gives the ratio of the equilibrium probabilities of populating each state: ‘‘ground’’ and ‘‘excited’’. As we have argued, the higher probability ground state is typically the one with higher density, i.e. the volume change  $v$  between the ground and the excited states is normally positive (see Fig. 3 for a schematic description of the model). Our two-state approximation is close in its spirit to recent Grinev–Edwards and de Gennes models [8,9].

We now introduce the concept of a base-line density,  $\rho_b$ , which corresponds to all the elementary modes at their ground states. Obviously, the experimentally–observed density is different from  $\rho_b$  due to a non-zero fraction of excited states:

$$\rho = \rho_b(t) \left( 1 - \frac{1}{V} \sum_n v^{(n)} \left( 1 + \frac{\kappa_{g \rightarrow e}^{(n)}}{\kappa_{e \rightarrow g}^{(n)}} \right)^{-1} \right). \quad (1)$$

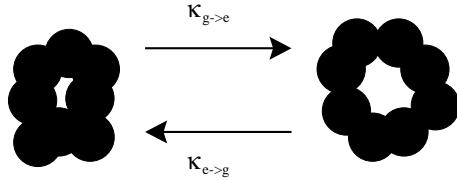


FIG. 3. Schematic description of the two-state model: at every tap a transition can occur from the ground to the excited state or vice versa.

The summation here is performed over all the domi-

nant two-state modes,  $V$  is the total volume and  $v^{(n)}$  is the volume difference between the excited and the ground  $n$ -th state. Assuming that the vibration intensity  $\Gamma$  is a qualitative analog of temperature, we expect the population of the excited states,  $P(\Gamma) = (1 + \kappa_{g \rightarrow e}^{(n)} / \kappa_{e \rightarrow g}^{(n)})^{-1}$ , to grow with  $\Gamma$ , starting from zero at  $\Gamma = 0$ . Hence, for a given  $\rho_b$ , the total density  $\rho$  will be lower at higher acceleration. This explains the observed effect of an abrupt change of  $\Gamma$ . After a switch from  $\Gamma_1$  to  $\Gamma_2$  at time  $t_0 = 0$ , the flip-flop mode contribution to the total density would relax to its new equilibrium value in the following way:

$$\Delta_{\Gamma_1 \Gamma_2}(t) = \rho_b \int v F_{\Gamma_1, \Gamma_2}(v, \kappa) (1 - \exp(-\kappa t)) dv d\kappa \quad (2)$$

Here  $\kappa$  is the relaxation rate of an individual mode, and the distribution function  $F_{\Gamma_1, \Gamma_2}(v, \kappa)$  is introduced as follows:

$$F_{\Gamma_1, \Gamma_2}(v, \kappa) \equiv \frac{1}{V} \sum_n \left( P^{(n)}(\Gamma_2) - P^{(n)}(\Gamma_1) \right) \delta(v - v^{(n)}) \delta \left( \kappa - \kappa_{g \rightarrow e}^{(n)}(\Gamma_2) - \kappa_{e \rightarrow g}^{(n)}(\Gamma_2) \right). \quad (3)$$

Since the observed density changes in compaction experiment are normally less than 1% of the total density [2,3], one can estimate the typical separation between neighboring flip-flop systems as 5 particle sizes, which yields a good justification for our no-coupling approximation. According to Eq. (2), if  $F_{\Gamma_1, \Gamma_2}$  does not vanish in the limit  $\kappa \rightarrow 0$ , the late stage of the relaxation of  $\Delta_{\Gamma_1 \Gamma_2}(t)$  is given by the power law:

$$\begin{aligned} \delta_{\Gamma_1, \Gamma_2}(t) &\equiv \Delta_{\Gamma_1 \Gamma_2}(t) - \Delta_{\Gamma_1 \Gamma_2}(\infty) = \\ &\rho_b \int v F_{\Gamma_1, \Gamma_2}(v, \kappa) e^{-\kappa t} dv d\kappa \sim \frac{1}{t}. \end{aligned} \quad (4)$$

Note that  $\rho_b$  is also dependent on time: although this cannot be described within our two-state approximation, the collection of elementary modes slowly evolves. Thus, one can observe two different processes: on short time scales, a fast relaxation due to the flip-flop modes is dominant, while over the long times, the dynamics are determined by the logarithmically slow evolution of the baseline density  $\rho_b(t)$ . The crossover between the two regimes is particularly obvious in Fig. 1b, where it results in a non-monotonic evolution. For experiments performed at sufficiently late stages of the density relaxation, the dynamics of the baseline density could be neglected compared to the contribution of the flip-flop modes (note that what we call a late-stage relaxation corresponds in fact to mesoscopic time scales which are always shorter than the relaxation time for  $\rho_b$ ). It has to be emphasized that the described experiments provide us with a tool for study of the response of the system, which is *not limited* to the nearly-equilibrium regime.

One can use our simple model to predict the response of the system to a more complicated pattern of changes of

$\Gamma$ . First, we reach, using annealing dynamics, a “quasi-steady” state at amplitude  $\Gamma_0$ , so that one can consider  $\rho_b$  constant later on. Let us switch the shaking acceleration from  $\Gamma_0$  to  $\Gamma_1$  for a finite number of taps  $\delta t$ , and then switch it back to  $\Gamma_0$ . During the intermediate  $\Gamma_1$ -stage, the system does not have enough time to completely relax to its new equilibrium. In our two-state model, the modes whose relaxation rate (at  $\Gamma_1$ ) is below  $\delta t^{-1}$  remain unrelaxed. Assuming that the slow modes at  $\Gamma_1$  are mostly the same as at  $\Gamma_0$ , we can calculate the backward density relaxation similarly to Eq. (4), with  $F(v, \kappa)$  effectively depleted below a minimal rate,  $\kappa_0$ . This cut-off frequency,  $\kappa_0$ , is expected to decrease monotonically with increasing perturbation duration  $\delta t$ . The resulting density relaxation after returning to  $\Gamma_0$  is given by:

$$\delta_{\Gamma_1, \Gamma_0}(t) \sim \frac{\exp(-\kappa_0 t)}{t}. \quad (5)$$

We tested these predictions by performing this three stage experiment, varying the duration,  $\delta t$ , of the perturbation( $\Gamma_1$ ) stage (Fig. 4). As predicted, the time needed to recover the steady-state density increases with the number of taps  $\delta t$  spent in the “hot” regime  $\Gamma_1 > \Gamma_0$ . In the coordinates chosen, the relaxation curves should follow the  $\delta t = \infty$  dynamics until the saturation at the cut-off time,  $\kappa_0^{-1}(\delta t)$ . We approximate the distribution function  $F$  by a constant above this low frequency cut-off at  $\kappa_0^{-1}(\delta t)$ , up to a high-frequency cut-off,  $\kappa_h \simeq 1 \text{ tap}^{-1}$ . This eliminates the unphysical low- $t$  divergence in Eq. (5). Figure 4 shows fits of the data to Eq. (2), where  $\kappa_0(\delta t)$  is determined from the fit. The best-fit is achieved at  $\kappa_h = 0.4$ , and the variation of this parameter would result in a simple rescaling of the time axis.

Figure 4 demonstrates good agreement between model and experiment, with some systematic error at the earliest relaxation stage (which is an expected result of our oversimplified description of the short time dynamics). For the late stage relaxation, we conclude that (i) within our experimental precision, the  $\delta t = \infty$  relaxation is consistent with the predicted  $1/t$  law; (ii) finite- $\delta t$  relaxation curves can be parameterized by a low frequency cut-off,  $\kappa_0$ ; and (iii)  $\kappa_0$  is a decreasing function of the waiting time  $\delta t$ , shown in the insert of the Figure 4. We now relate our picture to previous experimental and theoretical results. As discussed earlier, the wide range of relaxation times reveals itself both in our response measurements and in the fluctuation spectra of the density. It is tempting to relate these two kinds of data through an analog of a Fluctuation-Dissipation Theorem (FDT). Of course, there is no fundamental reason for FDT to be applicable to the granular system [10]. Even though the above two-state model could be mapped onto a thermal system (in which FDT is expected to work), the thermodynamic variable conjugate to density in the context of such a mapping has no clear physical meaning. Nev-

ertheless, below we outline the pseudo-FDT relationship expected for the granular system under rather natural approximation. Namely, we neglect the correlation between the volume change  $v$  and the life time  $\kappa^{-1}$  of an individual mode, i.e. assume  $F_{0,\Gamma}(v, \kappa) = f(\kappa)g(v)$ . Then the density autocorrelation function can be written as follows:

$$\langle \delta\rho(0)\delta\rho(t) \rangle_\Gamma = \frac{\rho^2}{2V} \int (\langle v^2 \rangle - \langle v \rangle^2) \exp(-\kappa t) f(\kappa) d\kappa = \frac{\langle v^2 \rangle - \langle v \rangle^2}{2\langle v \rangle V} \delta_{0,\Gamma}(t). \quad (6)$$

Thus, the density correlator is simply proportional to the response function corresponding to the switch between a very low acceleration (at which virtually all the modes are in their ground states) and the given one,  $\Gamma$ . An experimental check of this relationship requires further high-precision studies of both the relaxation dynamics and the fluctuation spectrum.

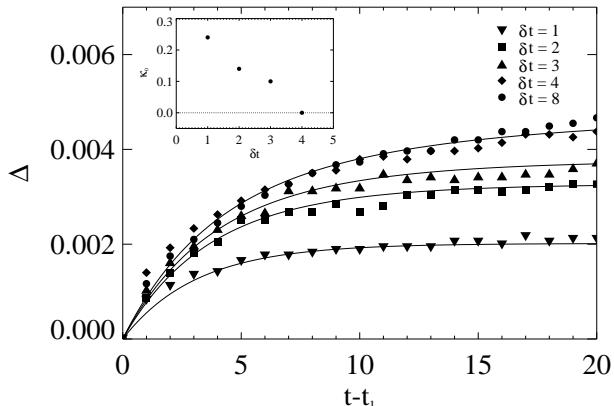


FIG. 4. The density relaxation ( $\Delta = \rho(t) - \rho(t_1)$ ) of the system, prepared by tapping for a long time at  $\Gamma_0 = 1.8$  and then tapping for a variable number,  $\delta t$ , of taps at a “hotter” intensity  $\Gamma_1 = 4.2$  before being returned to  $\Gamma_0$  at time  $t_1$ . Note that  $\rho(t_1)$  is a function of  $\delta t$ . Thus, while the asymptotic value of  $\Delta$  depends on  $\delta t$ , all curves asymptote to the same density  $\rho(\infty)$ , which depends only on  $\Gamma_0$  and not on  $\delta t$ . The solid lines represent the theoretical curves, with appropriate values of the parameter  $\kappa_0$ . The dependence of the cut-off rate  $\kappa_0$  on the waiting time  $\delta t$  is shown at the insert for  $\delta t \leq 4$  taps. We do not show the value for  $\delta t = 8$  since we found it null within the error bar, as for  $\delta t = 4$ . Each experimental graph is an average of 12 runs.

Our model also gives a simple interpretation to the decreasing dependence of the steady-state density on  $\Gamma$ : it can be attributed to the growth of the population of the excited states,  $P(\Gamma)$ . Indeed, the corresponding correction to the total density is about 1%, i.e. of the same order as the variation of the equilibrium packing fraction with  $\Gamma$  [3].

The slow dynamics associated with the evolution of the base-line density can also be addressed within our approach. For doing so we need to account for the

coupling of individual modes. Namely, it would be a reasonable assumption that a relaxation of one mode to its ground state may frustrate such a transition for some of its neighbors (e.g. in 3D the most compact local cluster can be created only at the expense of less dense neighboring regions). Thus, we arrive at an effective anti-ferromagnetic (AF) coupling (of an infinite strength) between the two-state modes. This extension of our model makes it remarkably similar to the so-called reversible Parking Lot Model (PLM) [11,12], which has been successful in describing many aspects of granular compaction experiments [2,3]. Recent simulations based on the “tetris model” [6] for compaction also find slow glassy responses to changes in  $\Gamma$ , but do not capture the short-term memory effect described here. [14].

In the PLM, D-dimensional space (the parking lot) gets packed with finite-size objects (cars), which may arrive and depart with fixed rates and which are not allowed to overlap. Now, the transcendental relationship between PLM and the granular compaction experiment is easier to explain: both PLM and our coupled flip-flop model belong to the same *generic* class of AF-type systems (in the case of PLM, a local two-state mode is represented by a particle whose center of mass may or may not be placed at point  $\mathbf{x}$ ; the mode coupling is due to the hard-core interactions). The PLM is known to capture the slow dynamics of granular compaction and some features of its fluctuation spectrum [3,12]. In fact, we performed numerical simulations of the PLM that display [13] the memory effects discussed in this work.

In conclusion, we used a sequence of abrupt switches of the shaking intensity  $\Gamma$  to study the response of a vibrated granular system. This technique can be used in the vicinity of the steady state density, as well as far from equilibrium. The major result is the direct demonstration of a memory effect in the system: the evolution is not predetermined by the local density alone, and its description requires introduction of additional “hidden” variables. Our phenomenological model for this behavior is built on minimal assumptions about the dynamics of the system and produces results which are generic and are expected to be valid for a wide class of more realistic microscopic models.

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